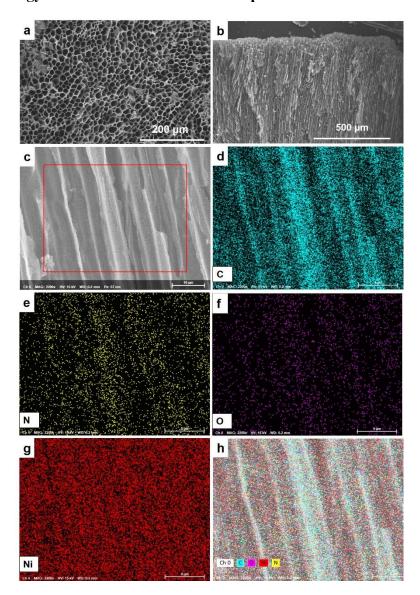
## Supplementary Materials for

Thickness-independent scalable high-performance Li-S batteries with high areal sulfur loading via electron-enriched carbon framework

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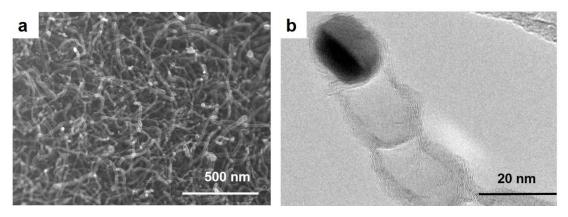
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<sup>b</sup> Materials Science and Engineering Program and Department of Mechanical
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### S1. Morphology characterization of wood-like precursor.



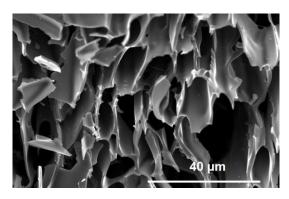
**Supplementary Figure S1.** Scanning electron microscope (SEM) images of (a) a cross-section and (b) a vertical section of precursor composite. (c) SEM image of precursor composite and the corresponding elemental mapping of (d) C, (e) N, (f) O, and (g) Ni.

### S2. Morphology characterization of N, O-CNTs.



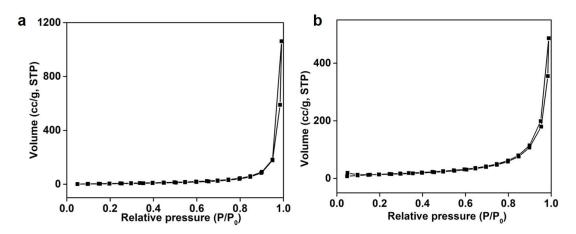
**Supplementary Figure S2.** (a, b) TEM images of CNONi-CNTs composite with tip growth of nickel particle.

## S3. Morphology characterization of WLC.



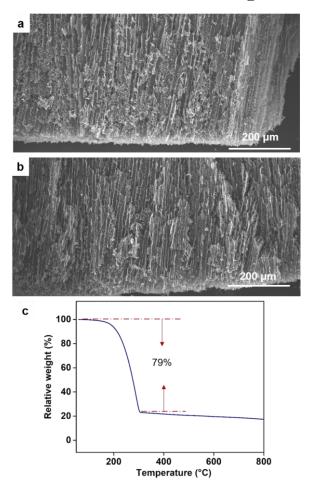
**Supplementary Figure S3.** SEM images of WLC.

### S4. N2 adsorption-desorption isotherms of WLC-CNTs and WLC.



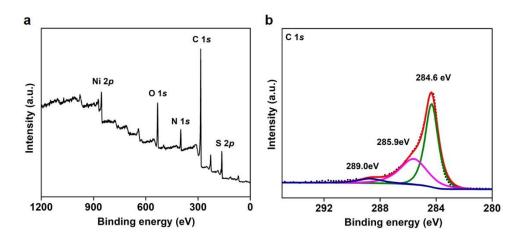
**Supplementary Figure S4.**  $N_2$  adsorption-desorption isotherms of (a) WLC-CNTs composite and (b) the bare WLC carbon framework.

### S5. Morphology characterization and TGA curve of S@WLC-CNTs composite.



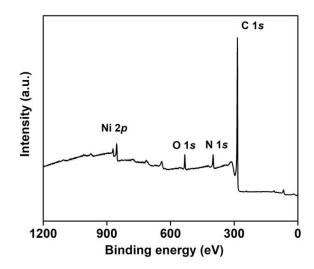
Supplementary Figure S5. (a, b) SEM images of selected different parts and (c) the TGA curve of the S@WLC-CNTs electrode with thickness of 400  $\mu m$ .

### S6. XPS characterization of S@WLC-CNTs composite.



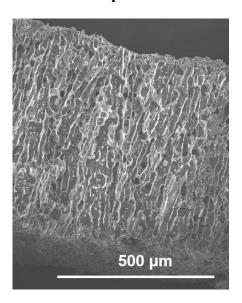
**Supplementary Figure S6.** (a) XPS survey spectrum of S@WLC-CNTs composite. (b) High-resolution XPS spectrum of C 1s.

### S7. XPS characterization of WLC-CNTs composite.



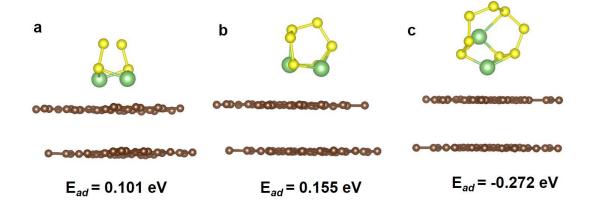
Supplementary Figure S7. XPS survey spectrum of WLC-CNTs composite.

### S8. Post-cycling of S@WLC-CNTs composite.



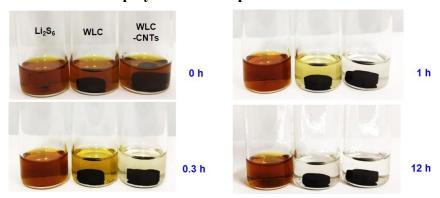
**Supplementary Figure S8.** SEM images of S@WLC-CNTs composite after rate cycling.

### S9. Adsorption energies of $Li_2S_n$ (n = 4, 6, 8) on the surface of carbon.



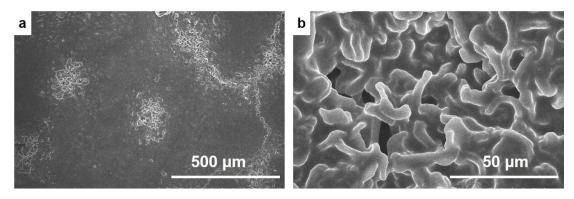
**Supplementary Figure S9.** Theoretical calculation of the adsorption energies of  $\text{Li}_2\text{S}_n$  (n = 4, 6, 8) on the surface of carbon.

### S10. Visual illustration of polysulfide entrapment.



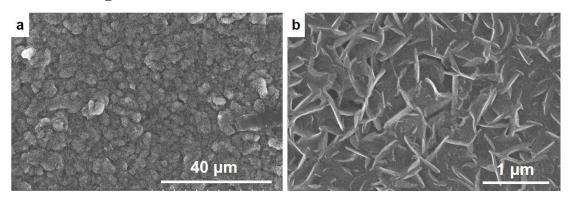
Supplementary Figure S10. Digital images of the sealed vials with  $\text{Li}_2\text{S}_6$  solution in DOL and DME during contact in the WLC, WLC-CNTs adsorption test.

### S11. SEM images of Li/Cu foil electrode.



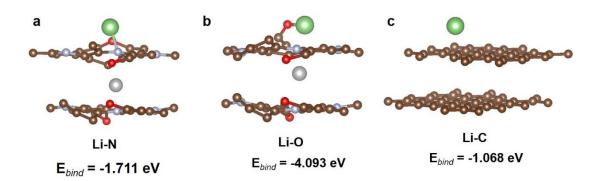
**Supplementary Figure S11.** SEM images of the planar Cu electrode with the amount of Li plating for areal capacity of 10 mAh cm<sup>-2</sup> at 1 mA cm<sup>-2</sup>.

### S12. SEM images of Li/WLC-CNTs electrode.



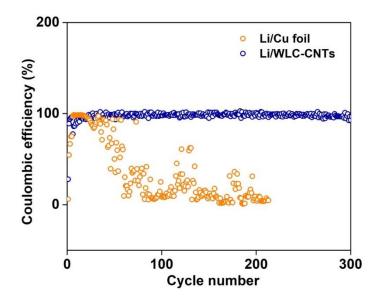
Supplementary Figure S12. SEM images of the top of the 400  $\mu m$  WLC-CNTs electrode with the amount of Li plating for areal capacity of 10 mAh cm<sup>-2</sup> at 1 mA cm<sup>-2</sup>

### S13. Modeling of Li binding energy.



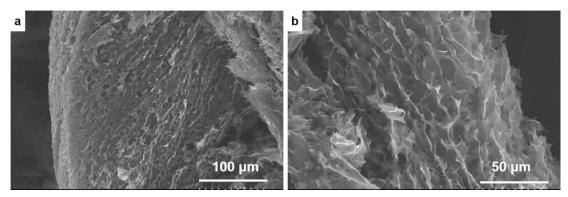
**Supplementary Figure S13.** Modeling of Li binding energy with (a, b) WLC-CNTs and (c) carbon surface (carbon is brown, lithium is green, oxygen is red, nitrogen is light blue, and nickel is grey between two carbon layers).

## S14. The Coulombic efficiency of Li/WLC-CNTs and Li/Cu electrodes.



**Supplementary Figure S14.** Comparison of the Coulombic efficiency of Li plating/stripping on the WLC-CNTs electrode and the planar Cu electrode with current density of 3 mA cm<sup>-2</sup> at fixed areal capacity of 3.5 mAh cm<sup>-2</sup>.

# S15. Post-cycling of Li/WLC-CNTs composite.



**Supplementary Figure S15.** SEM images of Li/WLC-CNTs composite after rate cycling.